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## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

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| <b>(51) International Patent Classification<sup>5</sup> :</b><br>A01N 37/18, 37/22, 37/24<br>A01N 25/26, 59/00, A61L 9/04  | <b>A1</b>   | <b>(11) International Publication Number:</b> WO 93/22919<br><b>(43) International Publication Date:</b> 25 November 1993 (25.11.93) |
| <b>(21) International Application Number:</b> PCT/US93/02186<br><b>(22) International Filing Date:</b> 16 March 1993 (16.03.93)<br><br><b>(30) Priority data:</b><br>881,691      12 May 1992 (12.05.92)      US<br><br><b>(71) Applicant:</b> CHURCH & DWIGHT COMPANY, INC.<br>[US/US]; 469 North Harrison Street, Princeton, NJ<br>08543 (US).<br><br><b>(72) Inventors:</b> WINSTON, Anthony, E. ; 42 Tall Oaks Drive,<br>East Brunswick, NJ 08816 (US). LAJOIE, M., Stephen ;<br>14 Launcelot Lane, Basking Ridge, NJ 07920 (US). JO-<br>SEPH, Amy, L. ; 323 Lambertville-Hopewell Rd., Hope-<br>well, NJ 08525 (US). JONES, Keith, A. ; 320 Flint Court<br>No., Yardley, PA 19067 (US). | <b>(74) Agents:</b> DEPAOLI, George, A. et al. ; 2231 Crystal Drive,<br>Suite 1103, Arlington, VA 22202 (US).<br><br><b>(81) Designated States:</b> AT, AU, BB, BG, BR, CA, CH, CZ,<br>DE, DK, ES, FI, GB, HU, JP, KP, KR, LK, LU, MG,<br>MN, MW, NL, NO, NZ, PL, PT, RO, RU, SD, SE, SK,<br>UA, European patent (AT, BE, CH, DE, DK, ES, FR,<br>GB, GR, IE, IT, LU, MC, NL, PT, SE), OAPI patent<br>(BF, BJ, CF, CG, CI, CM, GA, GN, ML, MR, SN, TD,<br>TG).<br><br><b>Published</b><br><i>With international search report.</i> |  |
| <b>(54) Title:</b> HERBICIDE COMPOSITIONS<br><br><b>(57) Abstract</b><br><br>This invention provides a novel herbicide composition having a bicarbonate-containing inorganic salt ingredient which enhances the efficacy of a herbicidal ingredient for weed control and plant growth regulation in agriculture and horticulture applications. An invention herbicide composition also contains a water-soluble organic compound which functions as a compatibility enhancing ingredient in aqueous formulations, and improves the spreadability and adhesiveness of the composition ingredients when applied to foliage.  |   |  |

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## HERBICIDE COMPOSITIONS

### BACKGROUND OF THE INVENTION

Weed control is essential in the cultivation of important agricultural crops such as corn, peanuts and cotton, and in the cultivation of many horticultural species. Also, the presence of weeds on non-cropped areas can be a fire hazard, or can result in undesirable drifting of sand or snow, or can cause discomfort to persons with allergies. Control of weeds is particularly beneficial when it permits the selective control of such plants without concurrent injury to desirable crops or vegetation.

The problems associated with effective control of undesired plants in varied agricultural and horticultural applications are significantly different. When treating crops or land which is prepared for crops, the herbicide chosen should be selective in its activity. It should kill weeds and other unwanted plants, and be harmless to the food crop plant growing or to be grown in the treated area. When treating certain other areas such as under power lines, it is desirable to kill all plant life, and a herbicide which is non-selective will be most useful in this application.

Chemical herbicides are classified generally according to the type of activity they possess. A given compound may have more than one type of activity depending upon its mode of

application and the rate at which it is applied.

In addition, herbicides are usually classified as selective or non-selective pre-emergents or post-emergents.

5           The most effective pre-emergence herbicide is one which is selective in its nature. If the designated compound can kill the seed and germinated seedlings of undesirable plants without harm to the seed and germinated seedlings of the crop, there  
10 will not be any problem of overpenetration of the soil by the herbicide.

          Post-emergence herbicides are applied after the crop and weeds have attained substantial height. In general, if a compound is found to have  
15 post-emergence activity it will not be selective. As a rule, herbicidally active compounds are not selective in their action on plant life.

          Of general interest with respect to the present invention are recent publications which  
20 describe newly developed herbicidal compositions. U.S. 5,022,917 discloses an extensive listing of herbicides that are effective through contact, and those that are taken up from the soil by root systems. The recited herbicides include defoliants,  
25 desiccants, eradicans, systemics and selective herbicides, and related plant growth regulants.

          Of particular interest with respect to the present invention are publications which disclose herbicide compositions which utilize a combination

of ingredients to achieve enhanced fungicidal activity. U.S. 4,508,559 describes a herbicide composition which contains 2-chloro- $\alpha,\alpha,\alpha$ -trifluoro-p-tolyl-3-carboxy-4-nitrophenyl ether and at least  
5 one water-soluble salt of an inorganic or organic acid, and which exhibits increased herbicidal efficacy because of the combination of ingredients.

There remains a continuing need for the development of new and more effective herbicides  
10 which can be applied to soil or plant foliage with a minimum of non-selective phytotoxic effects on cultivated crops.

Accordingly, it is an object of this invention to provide a biocide composition which is  
15 a blend of inorganic and organic compounds exhibiting properties suitable for weed eradication, or for growth control or defoliation applications with cultivated crops.

It is another object of this invention to  
20 provide a herbicide composition which is a dry blend of ingredients which include a bicarbonate salt which enhances the biocidal activity of a herbicide ingredient.

Other objects and advantages of the  
25 present invention shall become apparent from the accompanying description and examples.

DESCRIPTION OF THE INVENTION

One or more objects of the present invention are accomplished by the provision of a herbicide composition which is a dry blend  
5 formulation comprising (1) an ingredient selected from alkali metal and ammonium bicarbonates;  
(2) a compatibility enhancing ingredient selected from water-soluble organic compounds which are in solid form at a temperature below about 10°C; and  
10 (3) a herbicide ingredient.

In another embodiment this invention provides an aqueous herbicidal formulation having a content comprising (1) an ingredient selected from alkali metal and ammonium bicarbonates;  
15 (2) a compatibility enhancing ingredient selected from water-soluble organic compounds which are in solid form at a temperature below about 10°C; and  
(3) a herbicide ingredient.

The inorganic salt ingredient is selected  
20 from compounds which include sodium bicarbonate, potassium bicarbonate, lithium bicarbonate and ammonium bicarbonate. In a further embodiment, the inorganic salt ingredient can include an additional compound selected from sodium carbonate, potassium  
25 carbonate, lithium carbonate and ammonium carbonate.

The inorganic salt ingredient typically will comprise between about 10-80 weight percent, based on the weight of dry blend formulation.

Illustrative of inorganic salt ingredients in a formulation are sodium, potassium, lithium or ammonium bicarbonate, or mixtures such as sodium bicarbonate and potassium bicarbonate; sodium bicarbonate and ammonium bicarbonate; potassium bicarbonate and ammonium bicarbonate; sodium bicarbonate, potassium bicarbonate and ammonium bicarbonate; sodium bicarbonate and potassium carbonate; potassium bicarbonate and potassium carbonate; and the like.

Multiple inorganic salt compounds can be utilized in a broad range of molar quantities relative to each other. The molar quantity of a carbonate salt compound normally is determined by pH control considerations when aqueous formulations are prepared. The content of a carbonate salt compound can be varied to control the pH at a desired level in the range of 7.5-12. Aqueous herbicidal formulations of the present invention tend to have a higher herbicidal activity at higher pH values.

A compatibility enhancing ingredient of the present invention herbicide compositions is a water-soluble organic compound which is in solid form at a temperature below about 10°C. Suitable compounds include acetamide, acetylurea, alanine, aminoquanidine, aminomalonate salt, aminopyridine, arabinose, benzenesulfonate salt, benzoate salt, citraconate salt, citrate salt, crotonate salt,

cyclohexanol, dihydroxyacetone, dihydroxyacetone phosphate salt, dihydroxybenzene, dimethylurea, ethanolamine, ethyl alaninate, ethyl arsonate, ethylglycine, ethylurea, ethylenedisulfonate salt, 5 ethyleneurea, paraformaldehyde, fucose, glutamate salt, glycerol, glycerol nitrate, glycerol phosphate salt, glycogen, glycolic aldehyde, glyoxal, guanidine, hexamine, mannitol, fructose, glucose, hydroxyurea, lactate salt, lactose, lysine, maleic 10 amide, malonate salt, maltose, maltodextrin, methoxypyridine, methyl acetate, methyl carbamate, methyl ethyl sulfone, methyl glucoside, methylhydantoin, methylinositol, methylthiourea, methyluracil, methylurea, methylenedisulfonate salt, 15 muconate salt, naphtholdisulfonate salt, nitrobenzoate salt, nitropentanediol, nitrophenol salt, nitrourethane, pentaglycerol, phenol, phenylenediamine, polydextrose, propionamide, propyl carbamate, propylurea, purine, pyrazine, pyrimidine, 20 ribose, saccharate salt, sarcosinate salt, semicarbazide, sorbate salt, succinimide, sucrose, tartarate salt, tetrahydrobenzoate salt, tetrahydroquinoline, tetrazine, thiourea, threonine, triaminobenzene, triazole, triethylphosphine oxide, 25 triethylenetetramine, trihydroxybenzene, trimethylurea, urea, xlenol, xylose, xylylene glycol, polyvinylpyrrolidone, sodium carboxymethylcellulose, xanthan gum, guar gum, locust bean gum, gum acacia, gum tragacanth, 30 potassium alginate, potato agar, and the like.

The compatibility enhancing ingredient is incorporated in a quantity between about 0.5-20 weight percent, based on the weight of ingredients in a dry blend fungicide composition.

- 5 The term "water-soluble" as employed herein refers to a compatibility enhancing organic compound which has a solubility of at least about one gram per 100 grams of water at 25°C.

- 10 The herbicidal ingredient of an invention herbicide composition is included in a quantity which will provide a concentration between about 0.0001-10 weight percent of the medium which is being applied to seeds, plants, trees, soil, and the like.

- 15 The herbicidal ingredient can be selected from a wide variety of organic and inorganic compounds or mixtures which are known and used in agriculture and horticulture applications.

- 20 Amide herbicides are exemplified by commercial products such as Lasso and Dual, which are effective for pre-emergent or pre-planting applications.

- 25 Arsenical herbicides include cacodylic acid and the salts of monomethylarsinic acid and dimethylarsinic acid. Cacodylic acid is a defoliating or desiccating contact herbicide. Arsinic acid salts have lower contact toxicity and act through absorption.

Carbamate and thiocarbamate herbicides include Belanal, Betanex, Sutan, Eptam, and similar trademark products. These herbicides usually are applied to the soil and are taken up through the root systems.

Carboxylic acid herbicides are illustrated by commercial products such as Banvel, Garlon and 2,4-D. Various of these herbicides can be applied to the soil or to foliage, and are effective against broad leaf weeds.

Dinitroaniline herbicides include Balan and Treflan commercial products, which are applied to the soil to inhibit root growth and shoot growth, and exhibit low translocation.

Heterocyclic nitrogen-containing herbicides are illustrated by Aatrex, Basagran, Sencor and Velpar, which are applied to the soil for pre-emergent control.

Organophosphate compounds are useful as plant growth regulators and herbicides. This type of organic biocide structure is illustrated by Bensulide and Betasan.

Urea herbicides are nonselective and usually are soil applied. Urea-type commercial products include Lorox and Tupersan.

Quaternary herbicides include commercial products such as Avenge, Diquat and Paraquat, which have utility as contact foliar.

Other commercially available herbicides include ammonium sulfamate, Atrazine, Bentazon, Bromacil, Casoron, Chloroamben, Delapon, Diuron, Fluometuron, Glphosate, Linuron, Picloram, Trifluralin, sodium chlorate, sodium azide, and the like.

The types of weeds which are controlled by herbicide compositions include barnyard grass, green foxtail, wild oats, nightshade, velvetleaf, annual morningglory, yellow nutsedge, pigweed, downy brome, and the like.

Application of a present invention herbicide composition can be accomplished by employing conventional techniques and equipment. A herbicide composition can be a dry blend dusting powder or granulated product, or an aqueous spraying formulation.

Dusting powders are prepared by grinding together about 5-25 weight percent of the dry blend inorganic and organic herbicide composition ingredients with a solid diluent such as kaolin, talc, fuller's earth, wood flour, and the like.

Granules can be formed by impregnating pellets of filler with a fluid medium of the herbicide composition ingredients, or by pelleting a dry blend herbicide composition in admixture with a powdered filler.

An invention herbicide composition also can be in the form of a dispersible powder in combination with a surfactant to facilitate

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dispersion of the powder in an aqueous medium. The surfactant is incorporated in a herbicide composition in a quantity between about 1-20 weight percent, based on the weight of water-insoluble ingredients.

The surfactant can be a cationic, anionic or nonionic type, or a mixture thereof. Suitable surfactants include cetyltrimethylammonium bromide; sodium lauryl sulfate; sodium dodecylbenzene-sulfonate; ammonium lignosulfonate; condensation products of ethylene oxide with fatty alcohols, amines or alkylphenols; partial esters of fatty acids and hexitol anhydrides; and the like.

The ingredients in a invention herbicide composition can be selected to include nitrogen, phosphorus and potassium elements, in a ratio that allows the composition to function as a fertilizer in addition to its function as a herbicide, when applied to soil or cultivated crops.

An invention herbicide composition can include one or more other biologically active ingredients, such as those which exhibit fungicidal or insecticidal activity.

A herbicide composition of the present invention has a novel combination of properties for the practice of plant growth control in agricultural and horticultural applications.

A compound of the inorganic salt ingredient exhibits fungicidal properties when used in a small quantity (e.g., 100 ppm). If a larger

quantity (e.g., 300 ppm) is employed, the inorganic salt ingredient exhibits herbicidal properties, and the efficacy of the herbicide ingredient is enhanced by the presence of the inorganic salt ingredient. A  
5 lesser quantity of herbicide ingredient then can be employed to achieve a desired degree of plant growth control.

A present invention herbicide composition can be formulated to exhibit controlled  
10 phytotoxicity, or to minimize the toxic effects of salt stress on plants by the inorganic salt ingredient.

A significant feature of a present invention herbicide composition is the inclusion of  
15 a compatibility enhancing agent as an essential ingredient. Migration and settling of solid ingredients is minimized, and a dry blend formulation has a more uniformly distributed content because of the presence of the compatibility  
20 enhancing ingredient. An aqueous herbicidal formulation has exceptional long term stability, without phase separation and precipitation of solids.

As a further advantage, a present  
25 invention aqueous herbicidal formulation has improved spreadability and adhesiveness when applied to plant foliage, and resists post-application herbicide drift. An applied formulation also

exhibits humectant properties on coated foliage, and increased herbicidal efficacy because of the presence of the compatibility enhancing ingredient.

The following examples are further  
5 illustrative of the present invention. The components and specific ingredients are presented as being typical, and various modifications can be derived in view of the foregoing disclosure within the scope of the invention.

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EXAMPLE I

This Example illustrates the preparation of a herbicide dusting powder in accordance with the present invention.

5 A blend of the following ingredients is prepared:

|                       | <u>Parts</u> |
|-----------------------|--------------|
| KHCO <sub>3</sub>     | 30           |
| dichlobenil (Casoron) | 5            |
| 10 mannitol           | 5            |
| talc                  | 65           |

The formulated blend is milled to provide a powder with a particle size of less than 0.5 micron.

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EXAMPLE II

This Example illustrates the preparation of a dinitroaniline-containing herbicide composition.

5           A blend of the following ingredients is prepared as a wettable powder formulation:

|                           | <u>Parts</u> |
|---------------------------|--------------|
| NaHCO <sub>3</sub>        | 30           |
| benfluralin (Balan)       | 6            |
| 10       dihydroxybenzene | 2            |
| sodium lignosulfonate     | 2            |
| kaolin                    | 30           |

The blend is suspended in water to provide an aqueous formulation which contains about six  
15       pounds of benfluralin per 500 gallons of water.

The aqueous formulation is applied in a quantity providing two pounds of benfluralin per acre of turf grass for the control of crabgrass.

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EXAMPLE III

This Example illustrates the preparation of a granulated herbicide composition which has a high suspension capacity in water.

5 A mixture of the following ingredients is prepared:

|                       | <u>Parts</u> |
|-----------------------|--------------|
| alachlor (Lasso)      | 60           |
| NaHCO <sub>3</sub>    | 10           |
| 10 KHCO <sub>3</sub>  | 5            |
| hexamine              | 2            |
| sodium lignosulfonate | 20           |

The mixture is dispersed in water, and spray-dried at 180°C. The resulting granules have a  
15 70% suspension capacity in water.

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EXAMPLE IV

This Example illustrates the preparation of herbicide composition tablets which rapidly disintegrate and disperse in water.

|    |                             |              |
|----|-----------------------------|--------------|
| 5  |                             | <u>Parts</u> |
|    | diquat dibromide            | 40           |
|    | NaHCO <sub>3</sub>          | 35           |
|    | citric acid                 | 12           |
|    | Lomar PWA 10 <sup>(1)</sup> | 10           |
| 10 | glyoxal                     | 2            |
|    | sodium lignosulfonate       | 1            |

The ingredients are blended, and formed into tablets which disintegrate and disperse in water within about five minutes at 25°C.

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<sup>(1)</sup> sodium salt of alkylacylsulfonate condensation product (Jacques Wolf & Co.)

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EXAMPLE V

This Example illustrates the preparation of a water-dilutable liquid concentrate herbicide composition.

5 A liquid concentrate is prepared from the following ingredients:

|                                 | <u>Parts</u> |
|---------------------------------|--------------|
| lorazepam (Lorox)               | 10           |
| NaHCO <sub>3</sub> (300 mesh)   | 35           |
| 10 KHCO <sub>3</sub> (300 mesh) | 35           |
| oleic acid monoglyceride        | 30           |
| glyceryl monoctanoate           | 10           |
| glycerol                        | 5            |

15 The ingredients are admixed and heated at 40°C to form a concentrated liquid suspension. When the suspension is diluted with water, it forms a stable emulsion which has utility as a herbicide spray in agricultural applications.

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EXAMPLE VI

This Example illustrates the preparation of an aqueous herbicidal formulation stabilized with a nonionic surfactant phosphate ester salt.

|    |   |              |
|----|---|--------------|
| 5  |   | <u>Parts</u> |
|    | daminozide (Alar)   | 22           |
|    | KHCO <sub>3</sub>   | 10           |
|    | NH <sub>4</sub> HCO <sub>3</sub>  | 10           |
| 10 | polyoxyethylene<br>(7.6 mol)-<br>phenylphenol ether<br>phosphate diethanol-<br>amine salt | 5            |
|    | sucrose   | 5            |
| 15 | water   | 60           |

The formulation is sprayed directly on plant life as a growth regulating agent.

EXAMPLE VII

This Example illustrates the preparation of a herbicide-fertilizer composition.

A blend of the following ingredients is  
5 prepared:

|                                    | <u>Parts</u> |
|------------------------------------|--------------|
| melamine                           | 40           |
| urea                               | 30           |
| 10 potassium glycerol<br>phosphate | 20           |
| Atrazine (Aatrex)                  | 5            |
| KHCO <sub>3</sub>                  | 15           |

Granules are prepared by tumbling the  
blend, spraying added water to form tacky solids,  
15 and then drying the granulated product.

EXAMPLE VIII

This Example illustrates the effectiveness of a pre-emergence herbicide in accordance with the present invention.

5           An aqueous emulsion formulation is prepared with the following ingredients:

|                                     | <u>Parts</u> |
|-------------------------------------|--------------|
| hexazinone (Velpar)                 | 300          |
| $\text{NH}_4\text{HCO}_3$           | 10           |
| 10 $\text{K}_2\text{CO}_3$          | 20           |
| sorbitol                            | 5            |
| ethoxylated sorbitan<br>monolaurate | 30           |
| water                               | 95           |

15           The emulsion formulation is diluted with water to 250 ppm of herbicide ingredient. The diluted formulation is tested at the rate of 10 pounds per acre of herbicide ingredient, by drenching the formulation onto soil disposed in  
20       4.5 inch plastic pots which contain respectively weed seeds of velvet leaf, jimsonweed, tall morningglory, switchgrass, barnyard grass, and green foxtail.

25           The percent control of each weed type is determined two weeks after treatment in comparison with untreated controls. The results indicate essentially 100 percent control of each weed type germination.

WHAT IS CLAIMED IS:

1. A herbicide composition which is a dry blend formulation comprising (1) an ingredient selected from alkali metal and ammonium bicarbonates; (2) a compatibility enhancing ingredient selected from water-soluble organic compounds which are in solid form at a temperature below about 10°C; and (3) a herbicide ingredient.
2. A herbicide composition in accordance with claim 1 which additionally contains a surfactant ingredient.
3. A herbicide composition in accordance with claim 1 wherein the content of inorganic salt ingredient is between about 10-80 weight percent.
4. A herbicide composition in accordance with claim 1 wherein the content of inorganic salt ingredient comprises sodium bicarbonate.
5. A herbicide composition in accordance with claim 1 wherein the content of inorganic salt ingredient comprises potassium bicarbonate.
6. A herbicide composition in accordance with claim 1 wherein the content of inorganic salt ingredient comprises ammonium bicarbonate.

7. A herbicide composition in accordance with claim 1 wherein the content of inorganic salt ingredient comprises sodium bicarbonate and potassium bicarbonate.

5           8. A herbicide composition in accordance with claim 1 wherein the content of inorganic salt ingredient comprises sodium bicarbonate and ammonium bicarbonate.

10           9. A herbicide composition in accordance with claim 1 wherein the content of inorganic salt ingredient comprises potassium bicarbonate and ammonium bicarbonate.

15           10. A herbicide composition in accordance with claim 1 wherein the content of inorganic salt ingredient comprises sodium bicarbonate, potassium bicarbonate and ammonium bicarbonate.

11. A herbicide composition in accordance with claim 1 wherein the compatibility enhancing ingredient is a monohydroxy or polyhydroxy compound.

20           12. A herbicide composition in accordance with claim 1 wherein the compatibility enhancing ingredient is an amine derivative.

13. A herbicide composition in accordance with claim 1 wherein the compatibility enhancing ingredient is a carboxylic or sulfonic compound.

14. A herbicide composition in accordance with claim 1 wherein the compatibility enhancing ingredient is a phosphorus-containing compound.

15. A herbicide composition in accordance with claim 1 wherein the compatibility enhancing ingredient is a carbohydrate.

16. A herbicide composition in accordance with claim 1 wherein the compatibility enhancing ingredient is urea or a substituted urea.

17. A herbicide composition in accordance with claim 1 wherein the compatibility enhancing ingredient is a carboxylate or sulfonate salt.

18. A herbicide composition in accordance with claim 1 wherein the compatibility enhancing ingredient is a phosphate salt.

19. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is an amide herbicide.

20. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is an arsenical herbicide.

5 21. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is a carbamate or thiocarbamate herbicide.

22. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is a carboxylate herbicide.

10 23. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is a dinitroaniline derivative herbicide.

15 24. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is a heterocyclic nitrogen derivative herbicide.

25. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is a urea derivative herbicide.

20 26. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is a quaternary derivative herbicide.

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27. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is dichlobenil.

28. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is triclopyr.

29. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is benfluralin.

10 30. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is diquat dibromide.

15 31. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is alachlor.

32. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is lorazepam.

20 33. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is daminozide.

34. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is atrazine.

35. A herbicide composition in accordance with claim 1 wherein the herbicidal ingredient is hexazinone.

36. A herbicide composition in accordance with claim 1 which contains nitrogen, phosphorus and potassium elements in a ratio which is functional as a fertilizer formulation.

37. An aqueous herbicidal formulation having a content comprising (1) an ingredient selected from alkali metal and ammonium bicarbonates; (2) a compatibility enhancing ingredient selected from water-soluble organic compounds which are in solid form at a temperature below about 10°C; and (3) a herbicide ingredient.

38. An aqueous herbicidal formulation in accordance with claim 37 which additionally contains a surfactant ingredient.

39. An aqueous herbicidal formulation in accordance with claim 37 wherein the content of inorganic salt ingredient is between about 10-80 weight percent, based on the weight of ingredients.

## INTERNATIONAL SEARCH REPORT

PCT/US93/02186

**A. CLASSIFICATION OF SUBJECT MATTER**

IPC(5) : A01N 37/18, 37/22, 37/24, 25/26, 59/00; A61L 9/04

US CL : Please See Extra Sheet.

According to International Patent Classification (IPC) or to both national classification and IPC

**B. FIELDS SEARCHED**

Minimum documentation searched (classification system followed by classification symbols)

U.S. :

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

**C. DOCUMENTS CONSIDERED TO BE RELEVANT**

| Category*         | Citation of document, with indication, where appropriate, of the relevant passages  | Relevant to claim No.                 |
|-------------------|---|---------------------------------------|
| Y                 | US, A, 3,442,945 (OLIN) 06 May 1969, See columns 15-18.   | 1-19,31 & 37-39                       |
| Y                 | US, A, 3,937,730 (VOGEL ET AL.) 10 February 1976, See columns 12-14.  | 1-19,31 & 37-39                       |
| <del>X</del><br>Y | US, A, 4,933,000 (SOMLO) 12 June 1990, See claims 1-5 and 7, column 1, lines 13-32, column 6, lines 42-44, and column 7, line 16. | <u>1-5 &amp; 7</u><br>1-19,31 & 37-39 |
| Y                 | JP, A, 52/7438 (SAKAMOTO ET AL.) 20 January 1977, See the English abstract.   | 1-19,31 & 37-39                       |
| Y                 | CA, A, 2,013,918 (ZELLWEGER) 07 October 1990, See pp. 1-5, 7, 9 and 12.   | 1-19,31 & 37-39                       |

☒ Further documents are listed in the continuation of Box C.
 ☐ See patent family annex.

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| * Special categories of cited documents:  | *T  | later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention  |
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Date of the actual completion of the international search

11 MAY 1993

Date of mailing of the international search report

14 JUN 1993

 Name and mailing address of the ISA/US  
 Commissioner of Patents and Trademarks  
 Box PCT  
 Washington, D.C. 20231

Facsimile No. NOT APPLICABLE

Authorized officer

JOHN PAK

Telephone No. (703) 308-1235

 Ngutto Nguyen  
 NGUTTO NGUYEN  
 INTERNATIONAL DIVISION

## INTERNATIONAL SEARCH REPORT

International application N  
PCT/US93/02186

## C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

| Category* | Citation of document, with indication, where appropriate, of the relevant passages | Relevant to claim No. |
|-----------|--|-----------------------|
| Y         | US, A, 5,004,614 (STANIFORTH) 02 April 1991, See columns 5 and 9.                  | 1-19,31 & 37-39       |
| Y         | The Merck Index, 10th edition, Merck & Co., Inc., Rahway, NJ, 1983, pp. 818-19.    | 1-19,31 & 37-39       |

## A. CLASSIFICATION OF SUBJECT MATTER:

US CL :

504/121,123,334,335,336,337,338,339,340,341, &amp; 342; 71/DIG. 1; 424/44,421,715,716,717

## BOX II. OBSERVATIONS WHERE UNITY OF INVENTION WAS LACKING

This ISA found multiple inventions as follows:

Claims 1-39 lack unity of invention, and are drawn to the following distinct groups:

- I. Claims 19 and 31, drawn to compositions containing an amide herbicidal active ingredient, classified in 504/340+.
- II. Claim 20, drawn to compositions containing an arsenical herbicidal active ingredient, classified in 504/187 and 504/190.
- III. Claim 21, drawn to compositions containing a carbamate or thiocarbamate herbicidal active ingredient, classified in 504/300+.
- IV. Claim 22, drawn to compositions containing a carboxylate herbicidal active ingredient, classified in 504/313+.
- V. Claims 23 and 29, drawn to compositions containing a dinitroaniline herbicidal active ingredient, classified in 504/347.
- VI. Claim 28, drawn to compositions containing triclopyr, which is a pyridine moiety containing herbicidal active ingredient, classified in 504/254.
- VII. Claims 34-35, drawn to compositions containing atrazine or hexazinone, both of which are triazine derivatives, as the herbicidal active ingredient, classified in 504/227+.
- VIII. Claims 25 and 32, drawn to compositions containing an urea herbicidal active ingredient, classified in 504/327+.
- IX. Claim 26, drawn to compositions containing a quaternary derivative herbicidal active ingredient, classified in 504/345.
- X. Claim 27, drawn to compositions containing dichlorbenil as the herbicidal active ingredient, classified in 504/309.
- XI. Claim 30, drawn to compositions containing diquat dibromide, which contains a 6-member ring with two ring nitrogens, as the herbicidal active ingredient, classified in 504/235+.
- XII. Claim 33, drawn to compositions containing daminozide, which contains a carboxylic moiety as well as a "N-N" bond, classified in 504/320.
- XIII. Claim 36, drawn to fertilizer compositions, classified in 71/11+.

Claims 1-18 and 37-39 are readable on each of the invention Groups, and thus will be searched to the extent that they read on the elected Group(s). Claim 24 is readable on either Group VI or VII, and will be searched to the extent that it reads on Group VI or VII, if either of the Group is elected.

The above thirteen inventions are distinct, each from the others, because each of the inventions contains an active ingredient(s) or a class of active ingredients which are chemically and structurally distinct from the active ingredients of the other inventions. Each of the inventions have attained recognition in the art as a separate subject for inventive effort, and this is evidenced by the separate classifications of each of the inventions. Due to the diverse chemical structures of the different active ingredients, a search for one of the invention Group would require a different field of search than that required of another invention Group. Thus, the claims lack a unity of invention under PCT Rule 13.2 and 37 CFR 1.475.

# INTERNATIONAL SEARCH REPORT

International application No.  
PCT/US93/02186

## Box I Observations where certain claims were found unsearchable (Continuation of item 1 of first sheet)

This international report has not been established in respect of certain claims under Article 17(2)(a) for the following reasons:

1. ☐ Claims Nos.:  
because they relate to subject matter not required to be searched by this Authority, namely:
2. ☐ Claims Nos.:  
because they relate to parts of the international application that do not comply with the prescribed requirements to such an extent that no meaningful international search can be carried out, specifically:
3. ☐ Claims Nos.:  
because they are dependent claims and are not drafted in accordance with the second and third sentences of Rule 6.4(a).

## Box II Observations where unity of invention is lacking (Continuation of item 2 of first sheet)

This International Searching Authority found multiple inventions in this international application, as follows:  
Please See Extra Sheet.

1. ☐ As all required additional search fees were timely paid by the applicant, this international search report covers all searchable claims.
2. ☐ As all searchable claims could be searched without effort justifying an additional fee, this Authority did not invite payment of any additional fee.
3. ☐ As only some of the required additional search fees were timely paid by the applicant, this international search report covers only those claims for which fees were paid, specifically claims Nos.:
4. ☒ No required additional search fees were timely paid by the applicant. Consequently, this international search report is restricted to the invention first mentioned in the claims; it is covered by claims Nos.:  
1-18(PART), 19,31, AND 37-39(PART)

Remark on Protest

- ☐ The additional search fees were accompanied by the applicant's protest.  
☐ No protest accompanied the payment of additional search fees.